

Handbook UFIREG Project

Ultrafine Particles - an evidence based contribution to the development of regional and European environmental and health policy (2011 - 2014)

This project is implemented through the CENTRAL EUROPE Programme co-financed by the ERDF



Ultrafine Particles – an evidence based contribution to the development of regional and European environmental and health policy (UFIREG).

This document was prepared by the consortium of the UFIREG project. For more information, please visit the project website: www.ufireg-central.eu

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EXECUTIVE SUMMARY

Ultrafine particles - Invisible to our eyes, but detrimental to our health

Ultrafine particles (UFP) are the smallest constituents of airborne particulate matter: they are smaller than 0.1 micrometres and invisible to our eyes. Yet, their potential adverse effects on human health are of great concern because of their specific properties and acting mechanisms. Size governs the transport and removal of particles from the air and their deposition within the respiratory system and it is partly associated with the chemical composition and the source. UFP have little mass but high number and surface area concentration and a high content of elemental and organic carbon. Ambient UFP are built from gases or originate from combustion processes. In urban areas, they are emitted mostly by anthropogenic sources like traffic, domestic heating, and industrial processes.

Epidemiological studies have shown that particulate matter (PM) is associated with adverse health effects, especially in vulnerable population groups. Health effects of UFP are in part different from effects of larger particles such as $PM_{2.5}$ or PM_{10} as they deposit deeply in the lung and can for example penetrate the lung membranes, reach the bloodstream and are transported to different organs such as heart, liver, kidneys and brain. Evidence on short-term health effects of UFP, however, is still limited and no epidemiological studies of long-term exposures to ambient UFP have been conducted yet.

The UFIREG project

The project "Ultrafine particles – an evidence based contribution to the development of regional and European environmental and health policy" (UFIREG) started in July 2011 and ended in December 2014. It was implemented through the Central Europe Programme and co-financed by the ERDF. Five cities in four Central European countries participated in the study: Augsburg (Germany), Chernivtsi (Ukraine), Dresden (Germany), Ljubljana (Slovenia) and Prague (Czech Republic).

The aim of the UFIREG project was to improve the knowledge base on possible health effects of UFP and to raise overall awareness of environmental and health care authorities and the population. The project had two main areas of work: (1) exposure assessment to UFP and other air pollutants in the five European cities and (2) epidemiological studies assessing short-term effects of these particles on human cause-specific mortality and morbidity.

Exposure assessment

To investigate the exposure of the population to UFP, UFIREG partners have established standardised UFP measurements using custom-made mobility particle size spectrometers in the five project cities. The temporal variation of UFP was determined at one fixed monitoring site in each of the five project cities. Overall, the particle number concentration (PNC) of 10-100 nm particles varied between the five cities from May 2012 to April 2014. In summer, there was a considerable influence of new particle formation due to high global radiation and precursor gases, especially in Dresden and Prague. The results demonstrate that PNC in urban areas strongly depend on various factors such as the activity of different particle sources (traffic, domestic heating, long-range transport, etc.) whereby the everyday life of people plays an important role, meteorological conditions and cityscape. Overall, the project results showed that high quality measurements of size-resolved PNC and integration into routine monitoring networks are still a challenge.

Epidemiological assessment

Epidemiological studies in the frame of the UFIREG project have assessed the short-term effects of UFP on human mortality and morbidity, especially in relation to cardiovascular and respiratory diseases. Official statistics were used to determine the association between air pollution concentration and daily (cause-specific: respiratory and cardiovascular) hospital admissions and mortality. Associations of UFP levels and health effects were analysed for each city by use of Poisson regression models adjusting for a number of confounding factors. Results on morbidity and mortality effects of UFP were heterogeneous across the five European cities investigated. Overall, an increase in respiratory hospital admissions and mortality could be detected for increases in UFP concentrations. Results on cardiovascular health were less conclusive.

There is still (a) limited epidemiological evidence on the effect of short-term exposure to UFP on health; (b) insufficient understanding of whether the effects of UFP are independent of those of $PM_{2.5}$ and PM_{10} ; (c) no evidence on the effects of long-term exposure to UFP on health, and (d) little evidence showing which size ranges or chemical characteristics of UFP are most significant to health.

Discussion and conclusions

It is still not possible to draw definite conclusions about the specific health consequences of exposure to UFP. Further multi-centre studies such as UFIREG are needed preferably investigating several years in order to produce powerful results. There are still few epidemiological studies investigating adverse health effects of UFP.

Recommendations

The UFIREG project and other scientific studies have uncovered a number of knowledge gaps. The project consortium is suggesting further pursuing open research questions. Further research is needed to create more epidemiological evidence on the effect of short-term and long-term exposure to UFP on health, to increase the understanding of whether the effects of UFP are independent of those of $PM_{2.5}$ and PM_{10} , and to show which size ranges or chemical characteristics of UFP are most significant to health.

So far, no directives for the regulation of UFP in ambient air and almost no official measurement sites which routinely measure UFP exist. Even if further research is required to formulate guidelines or set threshold, policymakers and stakeholders are called upon for supporting routine measurements and research efforts as well as for developing and implementing measures to reduce UFP emissions, particularly from transport and domestic heating/biomass burning, at this stage.

1 BACKGROUND AND INTRODUCTION

Ultrafine particles (UFP) are defined as having a diameter of 100 nanometres (nm) and smaller. This means that the size of an ultrafine particle is about 1/1000 of the diameter of a human hair (Figure 1).

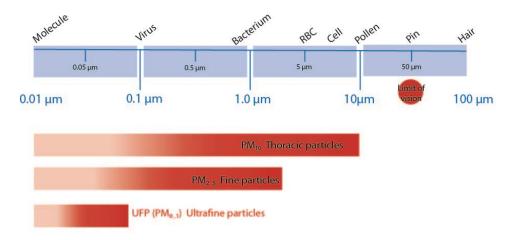


Figure 1. Size comparisons of particulate matter (adapted from Brook et al., 2004).

Ambient UFP sources are mostly related to combustion processes (e.g. traffic-related, industrial or from domestic heating). Motor vehicles are regarded as the leading source of UFP, especially in urban areas. Due to their small size and little mass the deposition and clearance of UFP in the respiratory tract is different compared to larger particles. Fine particles with an aerodynamic diameter < 10 μ m (PM₁₀) or <2.5 μ m (PM_{2.5}) deposit mainly in the upper and lower airways, whereas UFP can proceed deeply into the pulmonary alveoli (Figure 2).

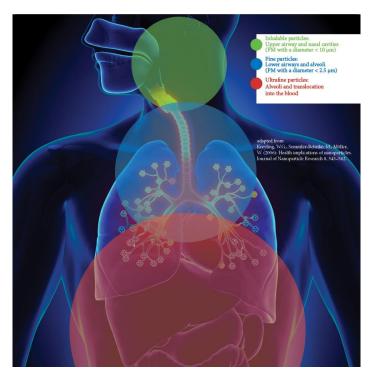


Figure 2. Regional deposition of particles in the human respiratory tract (adapted from Kreyling et al., 2006).

Epidemiological studies have shown that particulate matter (PM) is associated with adverse health effects (Rückerl et al., 2011) (Figure 3), especially in vulnerable population groups (Figure 4).

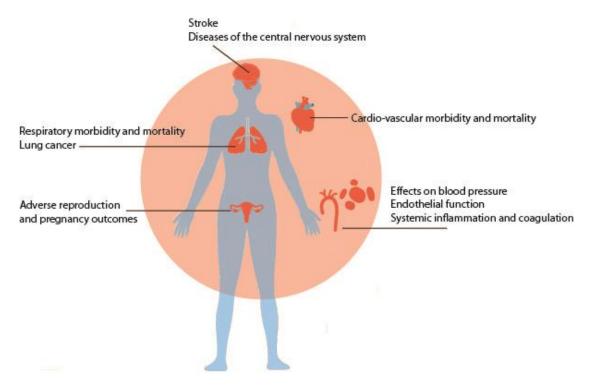


Figure 3. Organs of the human body that can be affected by air pollution (adapted from Peters et al., 2011).



Figure 4. Stages in life of special susceptibility toward air pollution (adapted from Rückerl et al., 2011).

Toxicological studies suggest that inhaled UFP can pass the lung epithelium and translocate into the blood to further be transported to other organs. Due to the differences in deposition and the potential for translocation as well as their huge active surface, it is assumed that effects of UFP differ and might be at least partly independent from those of larger particles such as PM_{10} and $PM_{2.5}$ (Box 1). So far, experimental studies do not provide sufficient evidence to confirm this hypothesis. Regarding health

effects, there is suggestive, but not consistent epidemiological evidence on the association between short-term exposures to UFP and cardiorespiratory health (WHO Regional Office for Europe, 2013; HEI Review Panel on Ultrafine Particles, 2013). Moreover, no epidemiological studies of long-term exposures to ambient UFP have been conducted yet.

Box 1. Ultrafine Particles (Rückerl et al., 2011; Brook et al., 2004; HEI Review Panel on Ultrafine Particles, 2013)

- Deposit deeply in the lung.
- Are not well recognized and cleared by the immune system in the alveolar space.
- Injure cells, cause oxidative stress, inflammation, mitochondrial exhaustion, and damage to protein and DNA.
- Penetrate the lung membranes, reach the bloodstream and can be transported to different organs such as heart, liver, kidneys and brain.
- Reach the brain via the olfactory nerve.

So far, no directives exist for the regulation of UFP measurements in ambient air. One of the conclusions of the WHO-REVIHAAP project (WHO Regional Office for Europe, 2013) was that the scientific base is too small to work on a guideline for the number of UFP and to propose a guideline value. Accordingly, in most locations UFP are not routinely monitored. However, scientists criticise that there are no limit values for UFP in ambient air and they argue that the established limit values for particulate matter (PM_{10} or $PM_{2.5}$) are not suitable for UFP because of their small size and mass. The most common measurement of UFP concentration is to count the number of particles per unit volume of air, which is typically referred to as particle number concentration (PNC). Thus, routine measurements of PNC and development of limit values could be important for better air quality control and for the prevention of potential adverse health effects.

Objectives of the UFIREG Project

The **main objective** of the UFIREG project was to assess UFP (and other air pollutant) concentrations in five European cities, to analyse their spatial and temporal variability, and to investigate the shortterm effects of ultrafine and fine particles on mortality and hospital admissions in order to provide an evidence based contribution to the development of environmental and health policy.

Moreover, the **specific aims** of the project were:

(1) to prepare a cost-efficient UFP measurement strategy applicable in all European regions,

(2) to demonstrate the applicability of the measurement strategy in the involved cities,

(3) to improve the overall sensitivity of the population as well as the environment and health care authorities of the targeted regions to potential adverse health effects of UPF,

(4) to integrate results into regional political decision-making and concrete local measures by developing differentiated solutions to the respective regional settings.

Project Description

The UFIREG project lasted from July 2011 to December 2014 and brought together a group of seven institutions from four European countries (Germany, Czech Republic, Slovenia and Ukraine), comprising experts in the fields of air pollution, epidemiology and public health. It was implemented

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through the CENTRAL EUROPE Programme co-financed by the ERDF.

The project aimed at improving the sensitivity of environment and health care authorities of targeted regions to UFP and to encourage closer co-operation between involved actors in order to better protect local populations from the negative health effects of exposure to UFP. Stakeholders from governmental agencies, regional authorities and policy, health care agencies, and science joined the project. The stakeholders were involved in order to integrate existing knowledge on UFP and information gained during the UFIREG project in regional air pollution management methodology, regional environment and health care policies.

The project partners implemented, harmonised and standardised PNC measurements in Augsburg and Dresden (Germany), Prague (Czech Republic), Ljubljana (Slovenia) and Chernivtsi (Ukraine) and investigated short-term effects of UFP on (cause-specific) mortality and hospital admissions in these cities. The results are intended to make a knowledge based contribution to the environmental policy in Europe (*Clean Air Plan* for Europe). It is worldwide one of the very few studies on short-term health effects of UFP which is based on multicentre time-series meta-analyses. The lack of epidemiological studies on UFP which follow this approach was clearly identified by the HEI report (2013) as one of the critical data gaps from the epidemiological perspectives. Furthermore, most research activities on UFP health effects were so far concentrated on Western European countries (or in USA).

For this study, measurements of PNC in different size ranges from 10 until 800 nm were conducted in Dresden and Augsburg since 2011, since 2012 in Prague and Ljubljana, and since 2013 in Chernivtsi (Figure 5). Due to the availability of mortality and hospital admission data as well as different start points of UFP measurement, the statistical analyses of health effects were conducted for the following periods: Dresden and Augsburg 2011-2012, Prague and Ljubljana 2012-2013 and Chernivtsi 2013.



Figure 5. Location of study areas and measurement sites.

2 METHODS

Measurements

In the scope of UFIREG, PNC measurements were performed using custom-made mobility particle size spectrometer, either Differential or Scanning Mobility Particle Sizer (DMPS/SMPS). They enable highly size-resolved PNC measurements in the range from 10 to 800 nm (except in Prague: 10 to 500 nm) with total number concentrations from 100 to 100,000 particles per cm³. Regular maintenance activities of the instruments and data processing as well as data validation were harmonised within the project. Data processing (so-called inversion) of the electrical mobility distribution (measured by the spectrometer) into the true particle number size distribution included the multiple charge correction according to Pfeifer et al. (2014), coincidence correction of the condensation particle counter (CPC) and the correction of the counting efficiency of the CPC. Particle losses due to diffusion in the inlet system and the spectrometer were also quantified using theoretical functions in the data evaluation software (Wiedensohler et al., 2012). The measurement principle and the data processing within UFIREG are described in a simplified form in Figure 6.

For comparable measurements between the cities, small uncertainties in sizing and concentration are required which can only be reached if the spectrometers are frequently quality assured. Therefore, an extensive quality assurance programme was an essential part of UFIREG aiming at a high data quality. It comprised staff training, an initial intercomparison of UFIREG spectrometers in a laboratory, frequent on-site comparisons against reference instruments, remote monitoring, and automated function control units at two sites.

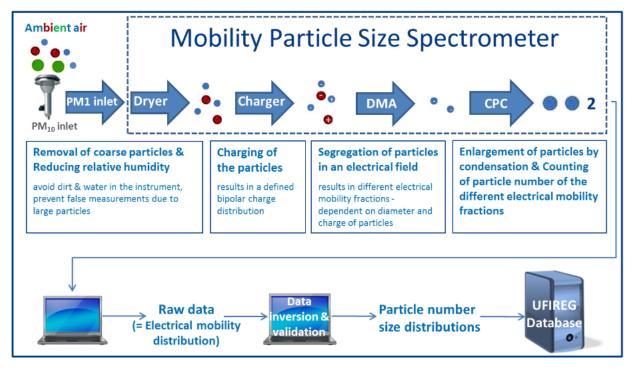


Figure 6. From particles in the air to particle number concentrations within the database: Simplified sketch of measurement principle and data processing within UFIREG (DMA: Differential Mobility Analyser, CPC: Condensation particle counter)

All of the UFIREG measurement stations were located at an urban or suburban background site which was representative for a large part of the urban population and had no roads with heavy traffic in immediate vicinity. Most of the measurement stations were integrated in local air quality monitoring

networks. At these sites, other air pollution parameters such as PM_{10} , $PM_{2.5}$, NO_x , SO_2 , O_3 , and partially black carbon as well as meteorological parameters were determined. The providers of the different pollutant and meteorological data are shown in Table 1. Since there were different regulations concerning air pollution and its monitoring in the Ukraine, only comparable PNC data could be used from Chernivtsi.

	Augsburg ¹	Chernivtsi	Dresden	Ljubljana ²	Prague
PNC	Helmholtz Zentrum München	L.I.Medved's Research Center of Preventive Toxicology, Food and Chemical Safety, Ministry of Health, Ukraine (State enterprise)	Saxon State Office for Environment, Agriculture and Geology	The National Laboratory of Health, Environment and Food	Institute of Chemical Process Fundamentals/ Czech Hydrometeorological Institute
PM/gas data	Bavarian Environment Agency	-	Saxon State Office for Environment, Agriculture and Geology	Slovenian Environment Agency	Czech Hydrometeorological Institute
Meteorology	Bavarian Environment Agency	www.hmn.ru/index.php ?index=8&value=33658	Saxon State Office for Environment, Agriculture and Geology	Slovenian Environment Agency	Czech Hydrometeorological Institute
EU station code for the official monitoring site	DEBY007/ DEBY099		DESN092	SI0003A	CZ0ASUC

Table 1. Sources of air pollutant data

Analysis of air quality data

For the comparative analysis of the air quality situation at the five UFIREG sites, a two-year dataset from May 2012 to April 2014 (Chernivtsi: January 2013 to April 2014) was considered.

The mobility particle size spectrometers operated within UFIREG deliver data in a 5 to 20 minute time-resolution. In general, hourly and daily averages were calculated with a threshold of 75% data availability. The overall availability of PNC data reached more than 75% at all stations (Table 2).

¹ Air pollutants were measured at different urban background stations within the city (3-4 km distance).

 $^{^{2}}$ Air pollutants were measured at different urban background stations within the city (600 m distance) until November 2013.

Table 2. Data availability of hourly PNC data in the UFIREG cities from May 2012 to April 2014(Chernivtsi: January 2013 to April 2014)

Augsburg	Chernivtsi	Dresden	Ljubljana	Prague
90%	86%	88%	77%	82%

Regarding temporal and spatial variation, the statistical analyses and result visualisation were carried out with the statistical open source project software R (R Core Team, 2014), and R package openair, (Carslaw, Ropkins, 2012a) and (Carslaw, Ropkins, 2012b).

To investigate the influence of air mass origin on particle number size distribution patterns, a meteorological cluster analysis based on back trajectories was performed³ according to the trajectoryclustering algorithm used before in Heintzenberg et al. (2011) which was principally based on the approach of Dorling et al. (1992). 48 hours back trajectories were computed using HYSPLIT, a trajectory model provided by the NOAA Air Resources Laboratory (Draxler, Hess, 2014) and radio soundings were obtained from national meteorological stations.

The frequency of the new particle formation (NPF) events was assessed⁴ using the method described in Dal Maso et al. (2005). Daily plots of particle number size distributions and daily plots of the positions of the modes of the multimodal distributions were considered. This method of the mode position determination using mathematical gnostics is described in Ždímal et al. (2008). Every day was classified into one of the category "event" (nucleation/new particle formation in other words), "non-event", "undefined", or "missing".

To define the impact of nucleation on the ultrafine particle number concentration, the nucleation strength factor was determined which was first introduced by Salma et al. (2014). It was calculated as the ratio of UFP and particles larger than 100 nm on nucleation days divided by the ratio of UFP and particles larger than 100 nm on non-event days. Undefined days were disregarded for this calculation.

Epidemiological data

On the basis of air quality data generated through UFIREG measurements, the project team investigated short-term effects of ambient air pollution on morbidity and mortality in the five cities involved in the project. Deaths statistics and hospital admission statistics were used for information on cause-specific morbidity and mortality.

Daily deaths and hospital admissions

Data on daily deaths and hospital admissions were collected for each of the five cities. Data on daily deaths were collected for the residents of the city who died in the city. Data on daily hospital admissions for each city were collected for the residents of the city who were hospitalised within the city. Only ordinary (no day-hospital) and acute (no scheduled) hospitalisations were considered, since the aim of the study was to investigate the association between daily pollutants concentrations and acute adverse health outcomes.

³ In cooperation with Leibniz Institute for Tropospheric Research.

⁴ In cooperation with Institute of Chemical Process Fundamentals of the ASCR.

The main diagnosis and cause of death, respectively, are based on the International Statistical Classification of Diseases and Related Health Problems (ICD-10). Deaths due to natural causes, deaths and hospital admissions due to cardiovascular and respiratory diseases were considered as primary outcomes. A list of all outcomes is shown in Table 3.

The study periods for the two German cities Augsburg and Dresden were 2011 to 2012, for Prague and Ljubljana 2012 to 2013 and for Chernivtsi 2013 only.

	ICD-10 code	Mortality	Hospital admissions
Natural causes of death	A00 - R99	Х	
Diabetes	E10 – E14		Х
Diseases of the circulatory system	I00 – I99	Х	Х
Cardiac diseases	I00 – I52	Х	Х
Ischaemic heart diseases	I00 – I25	Х	Х
Acute coronary events	I21 – I23	Х	Х
Arrhythmias	I46 – I49	Х	Х
Heart failure	150	Х	Х
Cerebrovascular diseases	I60 – I69	Х	Х
Haemorrhagic stroke	I60, I61		Х
Ischemic stroke	163, 165, 166		Х
Diseases of the respiratory system	J00 – J99	Х	Х
Lower respiratory tract infections (LRTI)	J09 – J18 + J20 – J22	Х	Х
Pneumonia	J12 – J18		Х
Chronic obstructive pulmonary disease (COPD)	J40 - J44 + J47	Х	Х
Asthma	J45 – J46		Х

Table 3. Outcomes including ICD-10 codes.

Confounder variables

We also collected information on other variables used for confounding adjustment. They include longterm time-trend (date order), indicator variables for weekdays and holidays, meteorological parameters (air temperature, relative humidity, barometric pressure) and – if available - influenza epidemics. Table 4 shows the providers of data on mortality, hospital admissions and influenza epidemics.

City	Mortality data	Hospital admissions data	Information on influenza epidemics
Augsburg	Research Data Centres of the Federal Statistical Office + Statistical Offices of the Länder	Research Data Centres of the Federal Statistical Office + Statistical Offices of the Länder	German Influenza Working Group of the Robert Koch Institute
Chernivtsi	Main Statistic Department in Chernivtsi Region	Collected directly from the hospitals	No data available
Dresden	Research Data Centres of the Federal Statistical Office + Statistical Offices of the Länder	Research Data Centres of the Federal Statistical Office + Statistical Offices of the Länder	German Influenza Working Group of the Robert Koch Institute
Ljubljana	National Institute of Public Health	Statistical Office of the Republic of Slovenia	National Institute of Public Health
Prague	Institute of Health Information and Statistics of the Czech Republic	Institute of Health Information and Statistics of the Czech Republic	National Institute of Public Health + Hygiene Station of the City of Prague

Table 4. Sources of health data.

Sociodemographic data

Sociodemographic data such as number of inhabitants (per age-group and per sex), estimated percentage of smokers, population density or number of newborns and deceased persons was used to describe the population in the cities involved in the project.

Data for Augsburg derived from the statistical yearbook of Augsburg. For Dresden data were obtained from the census in 2011 and the Statistical Office of the Free State of Saxony. The Statistical Office of the Republic of Slovenia provided sociodemographic data for Ljubljana. Data for Prague were obtained from the Institute of Health Information and Statistics of the Czech Republic and the Czech Statistical Office. For Chernivtsi data derived from the Main Statistic Department in Chernivtsi Region.

Epidemiological analysis

The association between air pollutants, and mortality or hospital admissions was investigated using Poisson regression models allowing for overdispersion. In a first step, a basic confounder model was set up a priori for all cities. This basic model included time-trend (date order), day of the week, holidays, the decrease of the populations present in the city during vacation periods, influenza epidemics (if available), air temperature, and relative humidity. In a second step air pollution effects were estimated by each city. The city-specific estimates were pooled using meta-analyses methods.

We performed single-lag models from lag 0 (same day of the event) up to lag 5 (five days prior to the event) to visually examine the lag structure of the association between particle exposures and health outcomes. Cumulative effect models were used to represent immediate effects (lag 0-1), delayed effects (lag 2-5) and prolonged effects (lag 0-5).

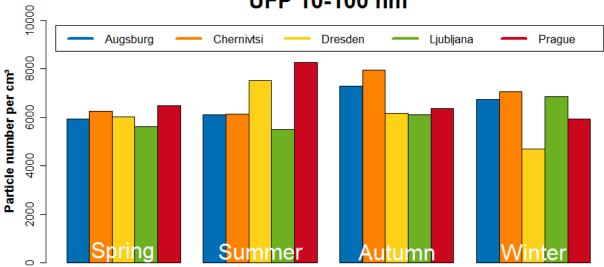
Cities were weighted according to the precision of the city-specific effect estimates. For pooling the

city-specific estimates the maximum likelihood effects estimator after van Houwelingen was used (van Houwelingen et al., 2002). We tested heterogeneity between the city-specific effect estimates using a Chi-squared test and by quantifying the degree of heterogeneity with the index I^2 . An I^2 of 50% indicates moderate heterogeneity and I^2 of at least 75% is an index for high heterogeneity between the city-specific estimates.

3 RESULTS

Temporal and spatial variation of PNC in the UFIREG cities

The temporal variation of PNC was determined at one fixed monitoring site in each of the five project cities. Overall, PNC and the temporal variation of different size fractions varied between the five sites. Figure 7 shows the seasonal variation of PNC between 10 and 100 nm from May 2012 to April 2014. The averaged PNC ranged from approximately 4,700 particles per cm³ in Dresden in the winter months (December to February) to 8,200 particles per cm³ in Prague in the summer months (June to August). In Prague and Dresden, there was a considerable influence of new particle formation due to high global radiation and precursor gases in summer. Leaf burning in autumn is still common in Chernivtsi and probably reflected by the high concentrations of particles shown in Figure 7 and 8. In winter, thermal inversion may lead to high PNC which was in particular typical for Ljubljana (Figure 7 and 8).



UFP 10-100 nm

Figure 7. Seasonal variation of particle number concentration (10-100 nm) in UFIREG cities from May 2012 to April 2014 (Chernivtsi: January 2013 – April 2014).

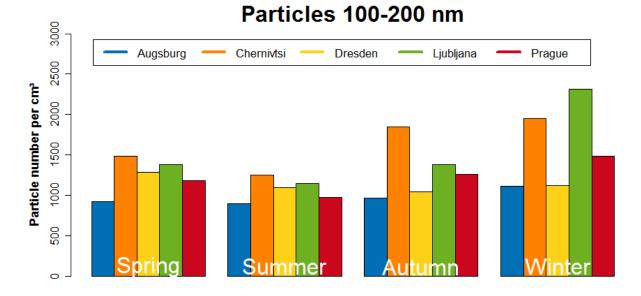


Figure 8. Seasonal variation of particle number concentration (100-200 nm) in UFIREG cities from May 2012 to April 2014 (Chernivtsi: January 2013 – April 2014).

In comparison to the PNC, the lowest PM_{10} concentrations during summer could be observed in Dresden which can be explained with different sources for UFP and PM_{10} and the negligible mass of nucleation particles which mainly cause the high PNC in Dresden in summer. As for PNC the variation between the cities was found to be smaller in spring but higher in winter. Due to inversion, the highest PM_{10} mass concentrations were also determined in Ljubljana in winter (Figure 9). Unfortunately, PM_{10} is not measured in Chernivtsi.

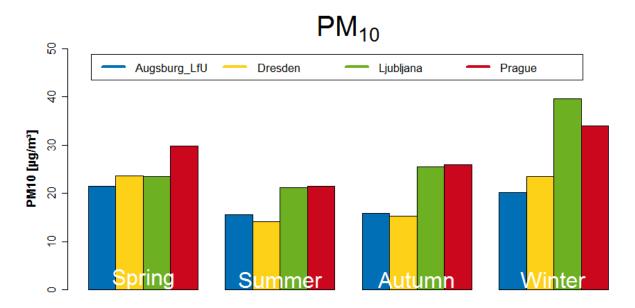


Figure 9. Seasonal variation of PM₁₀ in UFIREG cities from May 2012 to April 2014.

The season dependent weekly variation (Figure 10) shows the influence of different sources and everyday life of the citizens living around the different UFIREG sites. In spring, the PNC are more or less comparable during the working days at all UFIREG stations. At the weekends, PNC measured in Ljubljana and Augsburg decreased due to less traffic. Although these two stations fulfil the criteria for

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urban background classification, there is a clear traffic impact. In Dresden and Chernivtsi, there seems to be an active "particle producing" weekend life on Saturdays in summer but the overall highest averaged UFP concentrations could be observed in Chernivtsi on Saturdays in autumn. The increase in UFP concentrations in Ljubljana on Friday especially in autumn and winter could not be explained yet but was perhaps caused by commuters` traffic.

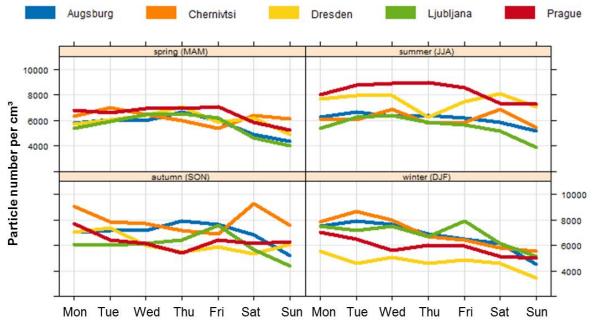
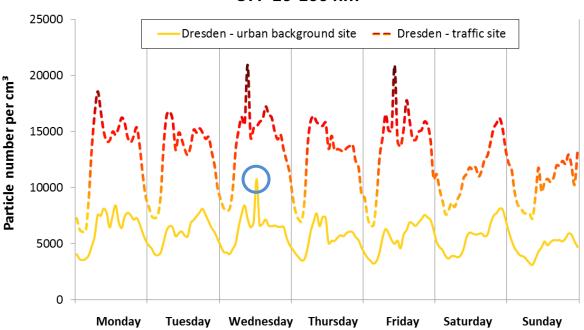


Figure 10. Weekly variation of particle number concentration (10-100 nm) in UFIREG cities from May 2012 to April 2014 (Chernivtsi: January 2013 – April 2014).



UFP 10-100 nm

Figure 11. Diurnal weekly variation for particle number concentration (10-100 nm) for two sites in Dresden in 2013; the urban background site is the UFIREG site; blue circle indicate maximum concentration caused by the operation of a leaf blower directly beside the station in October 2013.

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The averaged diurnal variation for each weekday is exemplarily shown for two sites in Dresden for the year 2013 in Figure 11. The UFP concentrations were about 1.5 times higher during the nights at the traffic site than at the background site. During the traffic rush hours, the difference was even more than twice as high. Moreover, the diurnal variation in Dresden showed differences between working days and weekend at both sites. During the nights on Friday and Saturday, in particular observed in summer, high concentrations were recorded probably due to barbecue and bonfires. In average, these "nightlife peaks" were almost as high as the traffic peaks during weekdays. The maximum peak on Wednesdays at the urban background station (marked with the blue circle) is caused by one single event – a leaf blower operated closely to the station in October 2013 which led to PNC over 1 million.

To assess the spatial variation between the UFIREG sites, the coefficient of divergence (COD) and the Spearman's rank correlation was calculated with daily means of size-fractioned PNC. The size dependency of the Spearman's rank correlation coefficient can be seen in Figure 12. The results indicate a strong relationship between PNC in Dresden and Prague and a slight to moderate relationship between Dresden-Augsburg and Prague-Augsburg with an increasing correlation coefficient for increasing particle sizes. For the PNC of all other UFIREG sites, almost no correlation could be found.

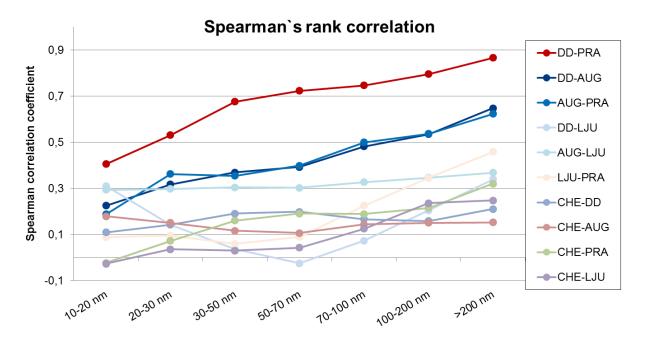


Figure 12. Spearman rank correlation of daily averages of all UFIREG cities from May 2012 to April 2014 (Chernivtsi: January 2013 – April 2014).

The link between PNC and concentrations of basic pollutants such as NO_2 , NO, SO_2 , PM_{10} and certain meteorological parameters was also examined within UFIREG in order to indicate whether and how the selected project stations are influenced by local sources and by different processes involved in the creation and growth of individual modes of particles in the air. Table 5 shows the results of Spearman's rank correlation between daily means of PNC and other air pollutants.

	10-20 nm	20-30 nm	30-50 nm	50-70 nm	70-100 nm	100-200 nm	>200 nm
\mathbf{PM}_{10}							
Augsburg_LfU ⁶	0,17	0,20	0,28	0,44	0,60	0,73	0,87
Augsburg_BP ⁷	0,06	0,10	0,19	0,38	0,56	0,66	0,82
Dresden	-0,12	0,07	0,21	0,44	0,65	0,81	0,93
Ljubljana	-0,02	0,17	0,30	0,47	0,65	0,80	0,88
Prague	-0,28	-0,05	0,18	0,48	0,70	0,84	0,92
NO_2							
Augsburg_LfU	0,59	0,65	0,73	0,77	0,80	0,79	0,53
Augsburg_BP	0,44	0,46	0,48	0,59	0,69	0,72	0,65
Dresden	0,08	0,21	0,22	0,33	0,46	0,55	0,64
Ljubljana	0,15	0,45	0,51	0,57	0,64	0,66	0,63
Prague	-0,16	0,07	0,17	0,42	0,64	0,74	0,77
NO							
Augsburg_LfU	0,57	0,64	0,72	0,79	0,82	0,79	0,52
Augsburg_BP	0,44	0,44	0,44	0,51	0,57	0,56	0,56
Dresden	0,13	0,23	0,23	0,29	0,37	0,43	0,49
Ljubljana	0,21	0,52	0,63	0,71	0,75	0,71	0,59
Prague	-0,03	0,17	0,25	0,44	0,61	0,66	0,64
SO_2							
Augsburg_LfU	0,15	0,15	0,17	0,22	0,26	0,28	0,25
Dresden	0,04	0,14	0,22	0,34	0,46	0,55	0,51
Ljubljana	-0,03	-0,02	-0,02	-0,01	0,04	0,10	0,12
Prague	-0,19	0,00	0,06	0,12	0,21	0,28	0,37

Table 5. Spearman's rank correlation coefficient between daily means of size-fractioned particle number concentrations and daily mean of PM₁₀, NO, NO₂ and SO₂ from May 2012 to April 2014⁵.

The correlation analyses indicate a different behaviour of the monitored stations. The stations in Ljubljana and in Augsburg were apparently more affected by traffic primary particle emissions than in Dresden and Prague as mentioned before. This was clearly reflected by correlation coefficients about 0.5 for correlation between the concentrations of the smallest particle fractions and nitrogen oxides in Ljubljana and Augsburg. The high correlation between NO_x and PNC in Augsburg is striking. The gaseous pollutants in Augsburg were measured not at the same location as PNC. The monitoring site at Bourges Platz is considered as urban background site and the measurement station at the premises of Bavarian Environment Agency even as regional background site. The high correlations between the traffic related air pollutants (PNC, NO, NO₂) measured at different urban background sites suggest that the single monitoring site operated in our study in Augsburg is representative for the entire city area. In Augsburg the traffic is the major source of locally generated particles. For Prague and Dresden, the correlation coefficient for this relation was smaller. At all stations, correlation analysis shows growing relation between PNC and other pollutants with growing particle size except for SO₂ which is weakly correlated with PNC, especially in Ljubljana.

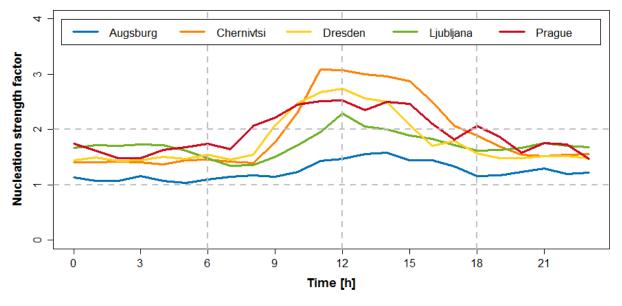
⁵ Graduation of the green colour from light to dark for a better visibility of correlation: coefficient R<0.2: no or only very weak relationship (light green); 0.2 < R < 0.5: weak to moderate relationship; 0.5 < R < 0.8: clear correlation; R>0.8: high or even perfect correlation (dark green).

⁶LfU: Bavarian Environment Agency.

⁷ BP: Bourges Platz.

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Another phenomenon which varied significantly between the UFIREG sites was new particle formation (NPF), a process mostly driven by high global radiation under certain conditions such as the concentration of precursor gases. The impact of NPF on PNC was assessed by calculating the nucleation strength factor. According to Salma et al. (2014), a value of the nucleation strength factor higher than two confirms NPF as the major source of UFP whereas a nucleation strength factor lower than one means other emission sources may contribute considerably to the overall UFP concentration. The daily cycle of the nucleation strength factor shown in Figure 13 emphasizes again the difference between particle sources at the sites in Ljubljana and Augsburg in comparison to the other stations. The contribution of NPF to the UFP concentration plays an important role in Dresden, Prague and Chernivtsi.



Influence of Nucleation

Figure 13. Diurnal variations of nucleation strength factor (NSF) at the UFIREG sites; the horizontal line at NSF value of 2 indicate when new particle formation influences PNC more than any other process.

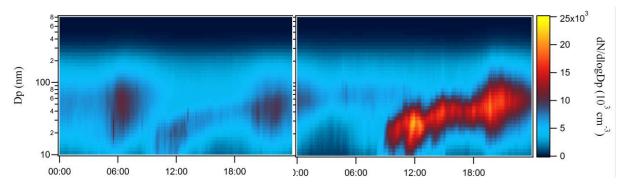


Figure 14. Averaged particle number size distribution of all summer days classified as new particle formation day; left: Ljubljana, right: Chernivtsi.

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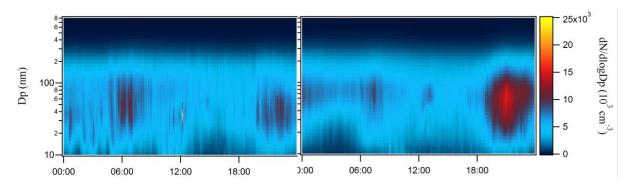


Figure 15. Averaged particle number size distribution of all summer days classified as non-event day; left: Ljubljana, right: Chernivtsi.

Figure 14 demonstrates the varying impact of NPF on the particle number size distribution in Ljubljana as example for almost negligible new particle formation and Chernivtsi where considerably more nucleation occurred resulting in a pronounced "nucleation banana". On average, particles grew up to about 50 to 60 nm in diameter on NPF days in all cities. Traffic influence can be seen at the Ljubljana site on NPF as well as non-event days, whereas in Chernivtsi fewer particles could be observed at the measurement site during the morning rush hour. However, high PNC in the evenings were always clearly visible in Chernivtsi (Figure 15).

Besides global radiation, other meteorological conditions favour high PNC. Therefore, a meteorological cluster analysis based on 48 hour back trajectories was performed within UFIREG which assessed the influence of air mass origin and thermal inversion or stability of stratification on particle number size distribution. The Figures 16 and 17 show the results exemplarily for Prague. In short, air masses coming from south and east, either warm (cluster 2 in red) or cold (cluster 1, 3, 4 in black/grey, green and blue), were characterised by higher number concentrations of particles larger than 100 nm if they reach Prague. In contrast, fast air masses from the west (cluster 7 in yellow) had apparently fewer particles.

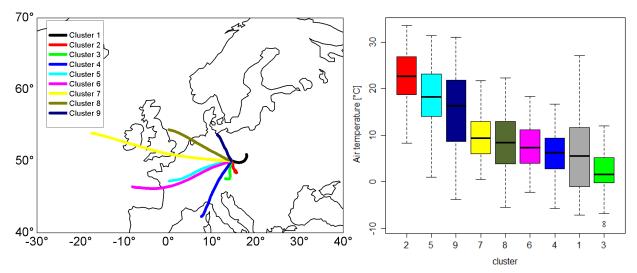


Figure 16. Left: Mean trajectories of nine air mass clusters coming to Prague; Right: Box plots of temperature for the nine clusters.

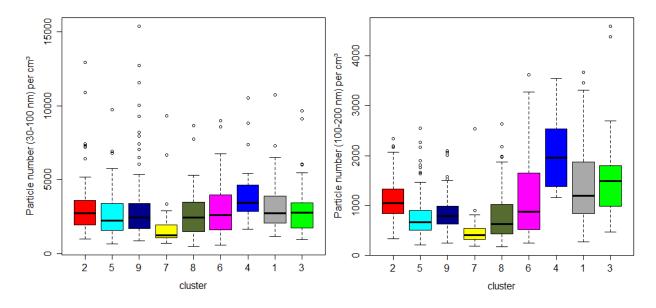


Figure 17. Box plots for particle number concentration in the range of 30-100 nm (left) and 100-200 nm (right) for the nine clusters.

In summary, different sources for UFP could be identified. They included different combustion processes such as from traffic, domestic heating, fireworks, bonfires, and leaf blowers. Also long-range transport and new particle formation contribute to UFP levels in the ambient air. The in-depth analysis of source apportionment for each site is still on-going and will provide important information concerning strategies for the improvement of urban air quality.

Descriptive statistics of health data

Table 6 shows a description of mortality outcomes by city for each year. In Augsburg, Dresden, Ljubljana and Prague 40%-50% of natural death cases were due to cardiovascular diseases. In Chernivtsi almost 70% of natural deaths were due to cardiovascular diseases in 2013.

City	Year	Population	Mean daily natural death counts (SD)	Mean daily cardiovascular death counts (SD)	Mean daily respiratory death counts (SD)
Augsburg	2011	266 647	6.9 (2.5)	3.1 (1.7)	0.5 (0.8)
	2012	272 699	7.2 (2.8)	3.1 (1.7)	0.4 (0.6)
Chernivtsi	2013	258 371	6.3 (2.7)	4.3 (2.1)	0.1 (0.4)
Dresden	2011	517 765	12.5 (3.6)	5.7 (2.4)	0.7 (0.9)
	2012	525 105	13.1 (3.8)	5.8 (2.5)	0.7 (0.9)
Ljubljana	2012	280 607	5.8 (2.5)	2.3 (1.5)	0.4 (0.6)
	2013	282 994	5.7 (2.4)	2.3 (1.5)	0.3 (0.5)
Prague	2012	1 246 780	27.1 (5.7)	13.7 (4.1)	1.5 (1.3)
	2013	1 243 201	26.5 (5.9)	12.8 (3.8)	1.7 (1.4)

Table 6. Description of (cause-specific)	mortality outcomes by city.
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Outcome definitions: natural causes ICD-10 A00-R99, cardiovascular diseases ICD-10 I00-I99, respiratory diseases ICD-10 J00-J99.

A description of hospital admissions by city for each year is shown in Table 7. So far, hospital admission data is available for Augsburg (2011-2012), Chernivtsi (2013), Dresden (2011-2012), Prague (2012-2013) and Ljubljana (2012). In all cities mean daily counts of cardiovascular hospital admissions were higher than the mean daily counts of respiratory hospital admissions.

City	Year	Population	Mean daily cardiovascular hospital admissions (SD)	Mean daily respiratory hospital admissions (SD)
Augsburg	2011	266 647	19.5 (8.5)	8.3 (3.9)
	2012	272 699	19.7 (8.8)	8.6 (4.4)
Chernivtsi	2013	258 371	12.1 (5.7)	5.2 (3.3)
Dresden	2011	517 765	34.0 (12.6)	11.7 (4.7)
	2012	525 105	34.3 (13.3)	11.5 (4.9)
Ljubljana	2012	280 607	14.4 (7.2)	8.2 (4.6)
Prague	2012	1 246 780	22.3 (8.7)	7.9 (4.0)
	2013	1 243 201	24.3 (8.1)	9.8 (4.8)

Table 7. Description of cause-specific hospital admissions by city.

Outcome definitions: cardiovascular diseases ICD-10 I00-I99, respiratory diseases ICD-10 J00-J99.

Effects of ultrafine and fine particles on natural and cause-specific mortality

For the analysis of UFP health effects, only the size range 20 to 100 nm was used due to the relatively high measurement uncertainty in the smaller size range 10 to 20 nm. Effects of UFP on mortality and hospital admissions are presented as percent changes in relative risk in association with each 1,000 particles/cm³ increase in daily UFP. Effects of $PM_{2.5}$ on mortality and hospital admissions are presented as percent changes in relative risk in association with each 10 µg/m³ increase in daily PM_{2.5}.

The pooled relative risk of natural mortality increased by 0.36% (95%-confidence interval:[-0.27, 1.00]) in association with a 1,000 particles/cm³ increase in daily UFP with a delay of one day (lag 1). However, the association was not statistically significant and no increase in natural mortality in association with an UFP increase could be found for other time lags.

Results on cardiovascular mortality were similar showing a delayed increase in the pooled relative risk with an increase in daily UFP concentration. The strongest effect was found with a delay of three days showing an increase in the risk of cardiovascular mortality by 0.33% [-0.57, 1.24] in association with an increase of 1,000 particles/cm³ in daily UFP. But overall, no significant increase in natural as well as cardiovascular mortality could be observed in association with an increase in UFP.

Chernivtsi was excluded from the analysis on respiratory mortality due to insufficient respiratory death cases in 2013. We observed an association between UFP and respiratory mortality across the remaining four cities. An increase in daily UFP concentration was associated with a 2.13% [-0.43, 4.75] increase in the risk of respiratory mortality with a five days delay (Figure 18).

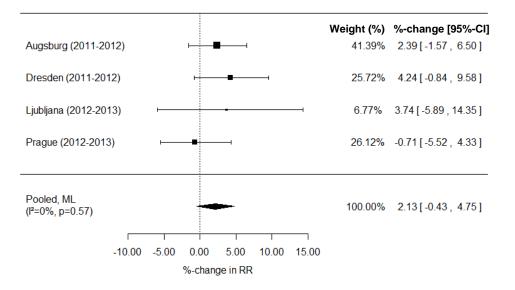


Figure 18. Percent change in respiratory mortality associated with each 1,000 particles/cm³ increase in daily UFP (lag 5).

Chernivtsi was excluded in the analyses on $PM_{2.5}$ since $PM_{2.5}$ was not available there. Effects of $PM_{2.5}$ on mortality were heterogeneous. Augsburg showed a significant increase in the relative risk of natural and cardiovascular mortality with increases in $PM_{2.5}$ with a delay of two to five days, whereas, Dresden showed decreases in the relative risk of natural and cardiovascular mortality. Non-significant but positive effect estimates were found for Prague and Ljubljana. Overall, the pooled effect estimates indicated a delayed increase in cardiovascular mortality in association with a 10 µg/m³ increase in $PM_{2.5}$ (Figure 19), but no effect of $PM_{2.5}$ on natural mortality. All cities showed non-significant associations between $PM_{2.5}$ and deaths due to respiratory diseases.

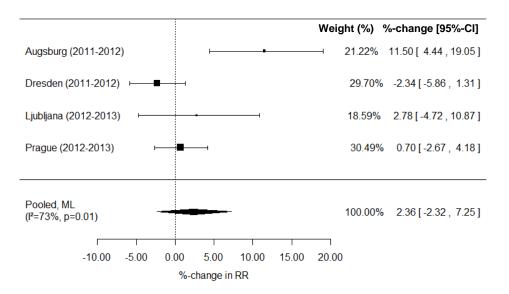


Figure 19. Percent change in cardiovascular mortality associated with each 10 μ g/m³ increase in daily PM_{2.5} (average of lag 2-5).

Effects of ultrafine and fine particles on cause-specific hospital admissions

Non-significant associations between UFP and hospital admissions due to cardiovascular diseases were found. But, we detected an increase in hospital admissions due to respiratory diseases in association with an increase in UFP concentration. The pooled relative risk increased significantly with an UFP increase in the 2-day average (1.44% [0.38, 2.53]) as well as with an UFP increase in the 6-day average (1.74% [0.35, 3.16]) (Figure 20).

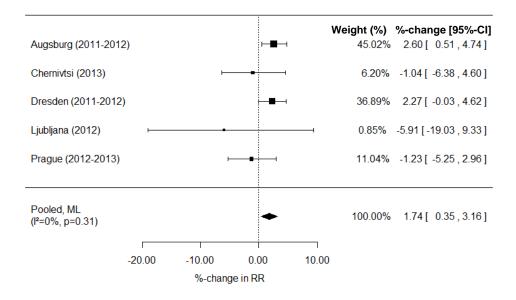


Figure 20. Percent change in respiratory hospital admissions associated with each 1,000 particles/cm³ increase in daily UFP (6-day average: lag 0-5).

We observed heterogeneous effects of $PM_{2.5}$ on cardiovascular hospital admissions. A 10 µg/m³ increase in $PM_{2.5}$ was associated with an increase in the relative risk of cardiovascular hospital admissions in Augsburg, Prague and Ljubljana. However, Dresden showed non-significant decreases in cardiovascular hospital admissions in association with an increase in $PM_{2.5}$ (Figure 21).

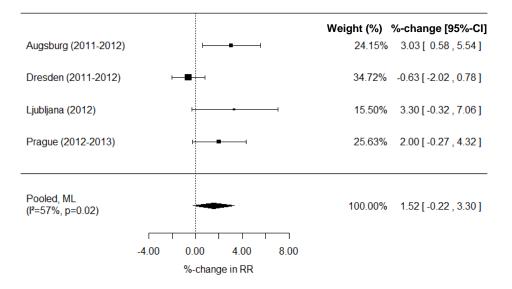


Figure 21. Percent change in cardiovascular hospital admissions associated with each 10 μ g/m³ increase in daily PM_{2.5} (lag 5).

The association between $PM_{2.5}$ and hospital admissions due to respiratory diseases was significant. The pooled relative risk of respiratory hospital admissions increased with a delay of one (3.06% [1.55, 4.60]) to five days (2.29% [0.60, 4.00]) as well as with an increase in the 6-day average of $PM_{2.5}$ (5.15% [3.00, 7.33]) (Figure 22).

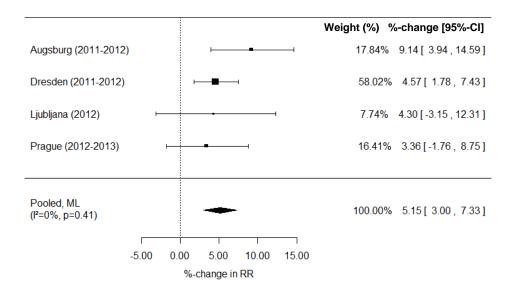


Figure 22. Percent change in respiratory hospital admissions associated with each 10 µg/m³ increase in daily PM_{2.5}(6-day average: lag 0-5).

4 DISCUSSION AND CONCLUSIONS

Quality assured measurements of UFP or of highly size-resolved PNC is still a challenge. However, within UFIREG the measuring routine could be improved considerably. Although an extensive quality assurance (QA) programme was performed within the scope of UFIREG, it emerged throughout the project that QA should be even more emphasised in future projects. To this end, important knowledge about future QA measures could be gained within UFIREG. The uncertainty of UFP/PNC measurements is still about +/- 20% due to the measurement principle in general and to the relatively new measurement technique and is, therewith, higher than for other air pollutants. Consequently, the instrument's quality and the measurement uncertainty need to be monitored and checked by reference instruments on-site on a very regular basis, especially with regard to epidemiological studies.

The complex QA, the high investment and running costs and the lack of legal regulations cause the sluggish integration of UFP/PNC measurements in official monitoring networks. In consequence, the data basis for epidemiological analyses is poor and epidemiological evidence for health effects of UFP is still not conclusive.

Determination of the total PNC by particle counters could be an alternative for the elaborate sizeresolved PNC measurements. However, only particle number size distribution data enable in-depth analysis, especially for identifying particle sources. These data are also more suitable for epidemiological studies such as in UFIREG.

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Following the comparative air quality analysis at the UFIREG sites, it revealed that PNC depends more on the special location of the measurement station (distance to the road, surrounding houses, traffic intensity, distance to the city centre, dominant wind direction) than it is the case for PM_{10} and $PM_{2.5}$. That should be considered if choosing the appropriate site for PNC measurements, especially for long-term epidemiological studies.

In summary, the results demonstrate that PNC in urban areas depend strongly on different factors such as the activity of different sources whereby the everyday life of people plays an important role, meteorological conditions, cityscape as well as orographic situation. The sources include different combustion processes such as from domestic heating, traffic, fireworks, bonfires or leaf blowers. Hence, a reduction of PNC is possible through less traffic, lower-emission vehicles, better air circulation in cities, less biomass burning (autumn and winter) and less bonfires/barbecues (summer).

The results of the epidemiological analyses indicate delayed effects of UFP on respiratory mortality as well as immediate and prolonged effects of UFP on respiratory hospital admissions. Moreover, results showed a significant association between $PM_{2.5}$ and respiratory hospital admissions. The effects of $PM_{2.5}$ on natural and cardiovascular mortality as well as cardiovascular hospital admissions were heterogeneous.

Further analyses regarding two-pollutant models and effect modification by several factors such as age, sex, season, wind direction and new particle formation are ongoing in order to find an explanation for the heterogeneity between the cities. Moreover, sensitivity analyses on the confounder models will be conducted to test the robustness of the results.

Due to different starting points of UFP measurements and because of delayed availability of health data in Germany it was not possible to use the same analysing periods for all five cities. Moreover, for Chernivtsi only one year could be investigated due to limited data availability. Differences in coding, difficulties in the exclusion of scheduled hospital admissions as well as in the restriction to people living in the city and hospitalised in the city might cause differences between the cities with regard to hospital admissions. However, UFIREG was one of the first multi-centre study investigating the effects of UFP on (cause-specific) mortality and hospital admissions including cities from Central and Eastern European countries since most research activities were so far concentrated on Western European countries. Moreover, it was the first study on UFP using harmonised UFP-measurements in all the five cities.

It is still not possible to draw definite conclusions on exposure to UFP and adverse health effects despite a growing scientific literature. Further multi-centre studies such as UFIREG are needed preferably investigating several years in order to produce powerful results. There are still few epidemiological studies investigating adverse health effects of UFP. Therefore, it is important to integrate UFP into routine measurement networks in order to provide data for short- as well as long-term epidemiological studies.

5 RECOMMENDATIONS

So far no directives for regulation of UFP in ambient air and almost no official measurement sites which routinely measure UFP exist. Usually, research results are used to formulate recommendations and guidelines, e.g. the WHO Air Quality Guidelines (WHO, 2005), which support policymakers in setting thresholds of air pollution constituents for national and European policy on air quality control

such as the EU Air Quality Directive⁸. At this stage, further research is required to draw definite conclusions on exposure to UFP and adverse health effects. Policymakers and stakeholders in turn can support routine measurements of UFP and research efforts at his stage.

Research

The UFIREG project and other scientific studies have uncovered a number of knowledge gaps: there is still (a) limited epidemiological evidence on the effect of short-term exposure to UFP on health; (b) insufficient understanding of whether the effects of UFP are independent of those of $PM_{2.5}$ and PM_{10} ; (c) no evidence on the effects of long-term exposure to UFP on health, and (d) little evidence showing which size ranges or chemical characteristics of UFP are most significant to health (WHO Regional Office for Europe, 2013).

Larger and more specific multi-centre studies and long study periods are needed to produce powerful results. The creation of so-called supersites or special sites should be considered (WHO Regional Office for Europe, 2013). Concentration–response functions need to be established for UFP and for newly identified health outcomes. This will also require the generation of large data sets on these exposure metrics (WHO Regional Office for Europe, 2013).

The project consortium is suggesting further pursuing open research questions (Box 2).

Box 2. Identified further research questions

- Are the short-term health effects of UFP comparable in cities across Europe? (More multicentre time-series studies including meta-analysis are needed.)
- What are the health effects of personal short-term exposures to UFP?
- What are the health effects of pollutant mixtures and together with individual activities i.e. in a tunnel or during physical activity?
- Are the health effects of UFP independent of the health effects of black carbon and/or other criteria air pollutants?
- What are the long-term health effects of UFP and their components?
- Are population groups spending more time near traffic more at risk compared to other groups?
- How effective are measures implemented for increasing air quality in urban settings?
- Which are the main sources of UFP and how to estimate the health effect impact of specific UFP sources?

Policymakers and stakeholders

As mentioned before, current data and studies on the levels of UFP and their health effects do not allow firm conclusions on exposure limits and respective health effects to be considered in European air quality guidelines. On the other hand, to date, UFP are not included in routine measurements of air quality monitoring stations. This in turn explains the lack of data for epidemiological studies. At this

⁸ Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe.

stage, policymakers and stakeholders are called upon for supporting routine measurements and research efforts to resolve this chicken-egg situation through:

- Continuing efforts to integrate UFP into routine measurement networks in order to provide data for short- as well as long-term epidemiological studies:
- Supporting multi-pollutant approaches, as so far pollutants are most often assessed independently.
- Fostering the conduct of epidemiological studies to assess the association between UFP levels and adverse health effects;
- Facilitating studies for evidence that may allow defining limit values for daily concentrations of UFP.
- Developing and implementing measures to reduce UFP emissions, particularly from transport and domestic heating/biomass burning. Measures may include:
 - ✓ Encouraging mass public transit and alternative energy sources for vehicles (electric and hybrid technologies);
 - ✓ Encouraging fewer road traffic journeys and more physically active transport;
 - ✓ Supporting concept of low emission zones (incl. shipping traffic in cities);
 - ✓ Supporting urban planning measures that help control hot spots such as near road microenvironments;
 - ✓ Supporting reliable filter systems for heavy duty vehicles (construction machinery), ships and heating systems.
- Helping protect people from UFP and soot particles also at occupational sites (e.g. construction sites).
- Strengthening communication and awareness raising for professionals and the public in relation to air pollution and particulate matter, including UFP.

High costs and lack of regulations could however hamper efforts to integrate UFP into routine measurements (Box 3).

Box 3. How much does UFP monitoring cost? (UFIREG, 2014)

Costs of particle number concentration measurements are composed of non-recurring investment costs, running costs (consumables, chemicals, electricity etc.), personnel costs and extraordinary costs. Investment and running costs have been estimated to be relatively high, on the basis of the UFIREG experience. High costs and lack of regulations prevent many institutions and authorities from including these measurements in the monitoring programmes of cities, regions or countries. UFIREG suggests re-evaluating future requirements and challenges of air quality control. Instruments to measure the distribution of particles sizes are expensive in investment and running costs and can only be introduced at special sites (supersites).

So far, no directives for the regulation of UFP in ambient air and almost no official measurement sites which routinely measure UFP exist. Even if further research is required to formulate guidelines or set threshold, policymakers and stakeholders can support the process at this stage.

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Box 3. How much does UFP monitoring cost?	

ULTRAFINE PARTICLES – AN EVIDENCE BASED CONTRIBUTION TO THE DEVELOPMENT OF REGIONAL AND EUROPEAN ENVIRONMENTAL AND HEALTH POLICY (UFIREG)



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NACIONALNI LABORATORIJ ZA ZDRAVJE, OKOLJE IN HRANO





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